Rapid HPGE Gamma Spectroscopic Mapping of Transuranic and High Energy Gamma Ray-Emitting Radionuclides for Remediation Planning

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ABSTRACT

Tank W-1A collected liquid wastes from several high radiation level facilities at the Oak Ridge National Laboratory. Previous environmental samples taken in the general area, particularly at Corehole 8, indicated radiological contamination in adjacent soil. The purpose of this characterization project was to generate three-dimensional maps of the extent of contamination in soil around the tank in order to estimate the volume of transuranic waste (TRU) and low level waste (LLW) for disposal and to plan the remediation operation.

A dynamic sampling plan was implemented in which results from each round of sampling determined which subsequent locations would be sampled. This plan maximized the value of data obtained while minimizing project costs and risk. To be effective, dynamic sampling requires that analytical results be available very rapidly. Geoprobe dual-tube soil sampling was used to collect soil cores up to 76 cm long. These cores were immediately brought to two adjacent high purity germanium (HPGe) gamma spectroscopy stations utilizing the *In Situ* Object Counting System (ISOCS) for quantitative analysis of gamma-emitting radionuclide activity levels. Typically, a 38-cm long core segment was measured for 15 minutes, and the results were reported within 30 minutes, satisfying the requirement for rapid turnaround times. The primary detected radioactive contaminants were ^{241}Am, ^{137}Cs, ^{152}Eu, ^{154}Eu, and daughters of ^{232}U, ^{233}U, and ^{232}Th.

The quantification of ^{241}Am in the presence of high concentrations of ^{137}Cs presented some special challenges since Compton effect from the 662 keV ^{137}Cs gamma rays raised the detection limits in the area of the characteristic 59.5 keV gamma line of ^{241}Am. This problem was solved using customized counting geometries, lead shielding, and collimators. It was possible to quantify 1.7 x 10^3 Bq/g of ^{241}Am in the presence of 7.8 x 10^5 Bq/g of ^{137}Cs. Detection limits for ^{241}Am were typically less than 370 Bq/g. Transuranic radionuclide detection limits as a function of ^{137}Cs activity concentration were compared for similar projects using geometries including 150 g sludge samples and 55 gallon waste drums.

TANK W-1A PROJECT BACKGROUND

Tank W-1A is a 2.3 m diameter x 4.1 m long underground storage tank in the North Tank Farm in the main plant area of Oak Ridge National Laboratory (ORNL). It was commissioned in 1951 to collect wastes from the nearby high-radiation-level facilities: Bldgs. 2026, 3019B, and the Radiochemical Processing Pilot Plant (Bldg. 3019). The liquid low-level waste (LLW) transfer line was suspected of leaking just above the tank inlet and the tank was taken out of service in 1986. Infiltrating groundwater was routinely discharged into the ORNL liquid LLW treatment

system. In 2000 the tank was acid cleaned and a video was taken of its bare metal internal surfaces. A Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) removal action in 2001 (Ref. 1) removed and dispositioned ~900 cubic meters of contaminated soil from three of the four quadrants around Tank W-1A. This purpose of this project was to characterize for disposition the LLW and TRU constituents in the remaining contaminated soils.

During the 2001 removal action eight grab samples of soil were collected and analyzed. These samples showed ^{241}Am in concentrations ranging from 2,000 to 11,000 Bq/g and $^{239/240}$Pu concentrations ranging from 400 to 52,000 Bq/g. In addition to the TRU isotopes, the soil samples contained other gamma-emitting radionuclides, including ^{137}Cs ranging from 44,000 Bq/g to 600,000 Bq/g. The high ^{137}Cs concentrations were the source of dose rates in the excavator bucket as high as 0.06 Sv/h. The removal action work was halted due to the unexpectedly high radiation levels found and the excavation was backfilled with clean soil.

FIELD GAMMA SPECTROSCOPY METHODOLOGY

One of the primary decision rules and drivers for this characterization project was the need to determine the spatial extent of the TRU (>3,700Bq/g) contamination in all three dimensions around the tank. It is anticipated that TRU-contaminated soils will eventually be dispositioned to the Waste Isolation Pilot Plant (WIPP) and LLW soils will be sent to the Environmental Management Waste Management Waste Facility (EWMWF) in Oak Ridge.

Analytical data from the prior removal action showed that the measured ^{241}Am concentrations in the eight grab samples scaled approximately with the total concentrations of all TRU radionuclides detected. The ^{241}Am to total TRU radionuclides ratio ranged from 0.18 to 0.74. The slope of the fitted linear curve describing this relationship has a slope of 0.21.

^{241}Am has a relatively high gamma yield and can be readily detected and quantified in soil samples in the field using high-resolution gamma spectroscopy. Other TRU waste constituents, in particular the Pu isotopes, have much lower gamma yields and are much more difficult to detect in the field. On this basis, high-resolution gamma spectroscopy was selected to provide rapid on-site quantification of ^{241}Am, ^{137}Cs, and other gamma-emitting radionuclides in soil cores as they were collected. A conservative scaling factor of 5.0 was applied to the ^{241}Am concentrations to estimate the total TRU concentrations. The estimated TRU and ^{137}Cs concentrations were mapped on a daily basis and used to guide the dynamic sampling plan.

Collection and Field Gamma Spectroscopy of Soil Cores

The overall sampling plan [3] was divided into three phases of work:

1. Phase 1 – systematic grid sampling
2. Phase 2 – build-out of grid to define the LLW/TRU boundary
3. Phase 3 – continued grid build-out to refine the LLW/TRU boundary and TRU core sampling

A small highly-maneuverable Geoprobe drill rig was used to collect soil cores at the grid locations. Drilling was performed from depths of 1.2 to 4.9 m. A DT21 sampling system

collected 2.85 cm-diameter soil cores up to 122 cm long into clear polyethylene terephthalate (PETG) tubes. These tubes were capped, wiped, and overpacked in 4.45 cm-diameter clear plastic tubes to prevent the spread of transferable contamination.

ISOCS gamma spectroscopy stations were set up onsite in two nearby trailers. A photo of one of the stations is shown in Fig. 1. Each station consisted of:

- a pair of PVC cradles to hold the soil cores
- 50 mm-thick lead shielding along the length of the cradles
- an ISOCS HPGe detector and liquid N_2 cryostat
- 50 mm-thick donut-shaped lead shields around the detector
- (optional) cone-shaped lead collimators in front of the detector
- additional lead brick shielding around the detector
- a heavy duty lift cart
- digital amplifier and laptop personal computer for data acquisition and analysis

The purpose of the extensive lead shielding was to allow for direct measurement of 30 cm-long segments of each core directly in front of the detector and blocking of gamma rays from other segments of the core being measured. The shielding was necessary due to the high concentrations of ^{137}Cs present in many of the soil cores.

Overpacked soil cores were brought one at a time to the measurement stations. When a core arrived it was first weighed on a digital scale so that the soil density could be calculated. Then it was placed in a cradle to the left of the HPGe detector. Thirty cm-long segments were marked along the length of the core and the core was then slid along the cradle so that the first segment was exposed in front of the detector. Acquisition of a gamma spectrum was then started. In most cases a 180-degree collimator was used around the detector and the sample-to-detector standoff distance was set at 15 cm. The standard counting time was 15 minutes per 30 cm segment.

Core segments with moderate to high concentrations of ^{137}Cs generated high gamma fluxes at the detector, leading to undesirably high deadtimes, distortion of the gamma spectra, and degraded detection limits. When the deadtime exceeded 20% the standoff distance was increased and, in some cases, additional lead collimation was added in front of the detector to reduce the field of view.

Gamma spectra were analyzed onsite immediately after they were collected. Unique ISOCS efficiency models were created for each specific combination of sample-detector geometry, ISOCS detector, and shielding/collimation. Table I lists the parameters that were incorporated and accounted for in each ISOCS model. After the ISOCS models were set up, the gamma spectra were analyzed for 25 project-specific radionuclides. A hardcopy NDA Report was then generated that included the measured activity, calculated uncertainty, and calculated detection limit for each analyte. A Transportation Report was also generated for each core segment to support shipping decisions. Reports for individual core segments were typically generated within 30 minutes of the start of measurements. Based on the NDA results, a small number of soil samples were shipped offsite for radiochemical analyses.

Fig. 1 Photo of one of the high-resolution gamma spectroscopy stations in a trailer at the Corehole 8 Tank W-1A site.

Table I: Sample/Geometry/Detector Parameters Included in ISOCS Efficiency Models

soil composition	sample diameter	attenuation in sample matrix
soil density	sample-detector distance	attenuation in sample walls
sample length	detector efficiency	attenuation in detector collimator

RESULTS OF FIELD GAMMA SPECTROSCOPY MEASUREMENTS

Approximately 300 soil core segments were measured in this campaign over eight weeks. The primary detected radioactive contaminants were ^{241}Am, ^{137}Cs, ^{152}Eu, ^{154}Eu, and daughters of ^{232}U, ^{233}U, and ^{232}Th. Pu isotope activities were below detection limits due to their very low gamma abundances and were not reported.

Fig. 2 shows a two-dimensional map of the measurement area with a 1m x 1m grid. The blue dots indicate the locations of the 64 probe holes and the black rectangle shows the approximate location of the buried tank. ^{241}Am was detected in 59 of the 64 holes. The measured ^{241}Am concentrations ranged from 2 to 17,612 Bq/g. The highest ^{241}Am concentrations occurred near

the northeast of the tank in the approximate location of the known historical leak in the input line to the tank. Detection limits for ^{241}Am were typically well below the project data quality objective of 370 Bq/g, which corresponds to one-tenth of the decision level for TRU waste. The soil core with the highest ^{137}Cs concentration of 788,000 Bq/g had a measured ^{241}Am concentration of 1,700 Bq/g.

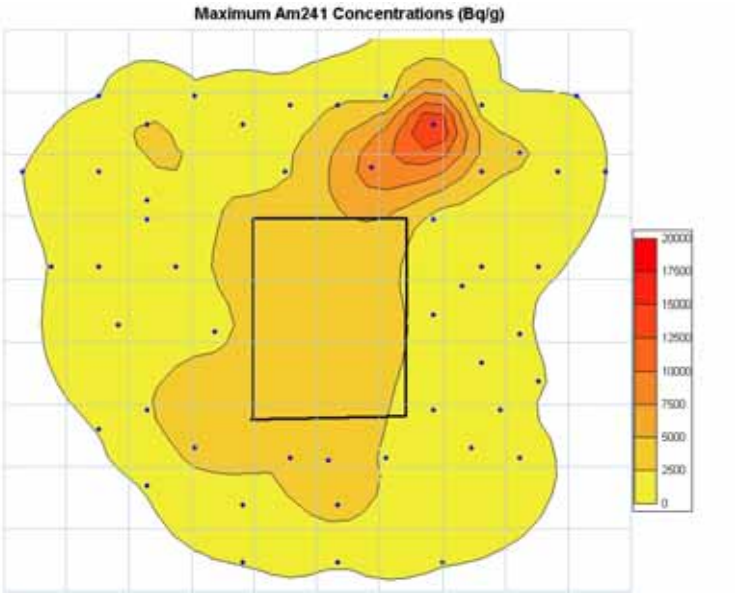


Fig. 2 Two dimensional map of interpolated maximum Am-241 concentrations.

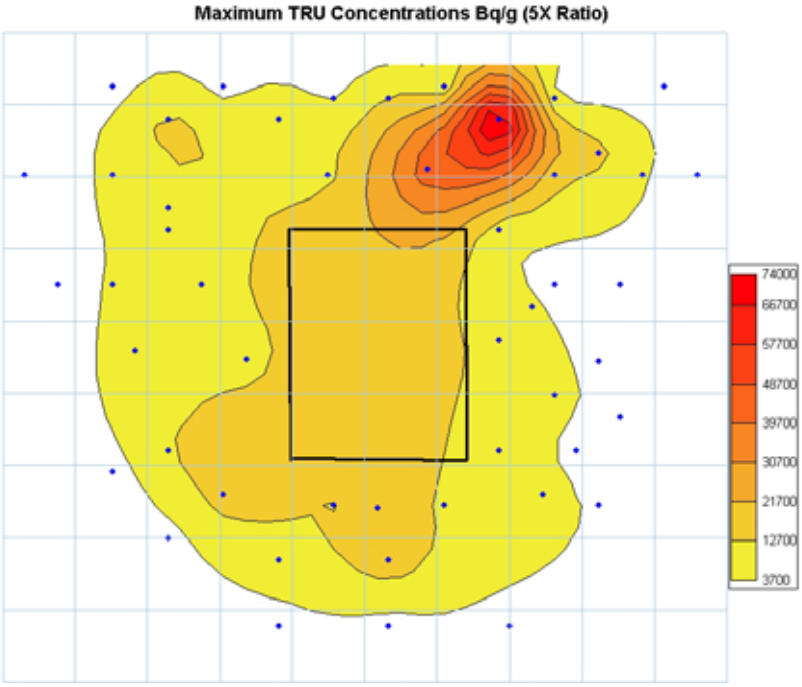


Fig. 3 Two dimensional map of maximum total TRU concentrations based on measured Am-241 concentrations and a scaling factor of 5.0.

When the scaling factor of 5.0 is applied to the maximum measured ^{241}Am concentrations, a contamination map of the extent of the maximum total TRU constituents can then be generated. This is shown in Fig. 3. The contour scale in this figure was set such that the outermost contour corresponds to the definition of TRU waste: 3700 Bq/g. Therefore some or all of the soil located within the outermost contour in Fig. 3 will need to be handled and dispositioned as TRU waste. Everything outside of that boundary will likely be handled and dispositioned as LLW.

COMPTON EFFECT FROM CS-137 AND DETECTION LIMITS FOR TRANSURANIC RADIONUCLIDES

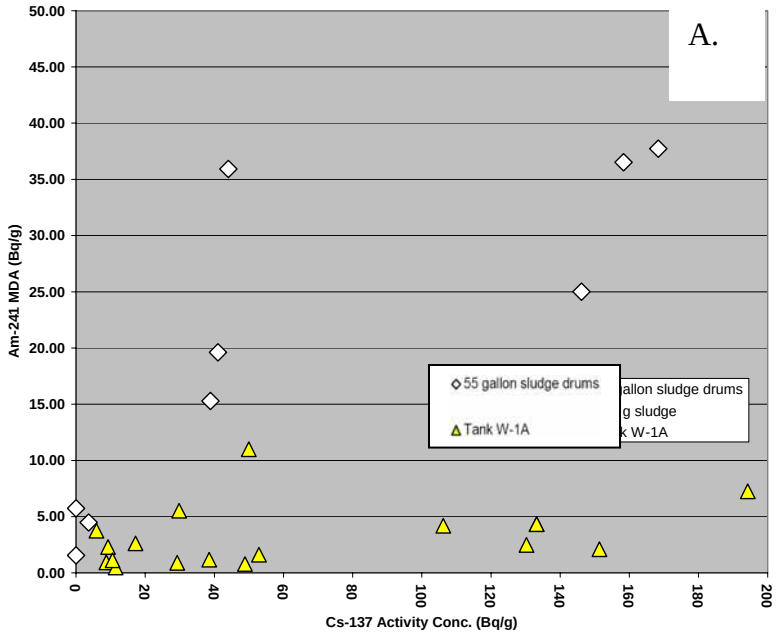
Compton effect from the high relative levels of ^{137}Cs in the samples was expected to significantly drive up HPGe detection limits for ^{241}Am. The 662 keV ^{137}Cs gamma ray raised the detection limits in the area of the characteristic 59.5 keV gamma line of ^{241}Am, despite precautions involving geometry, collimation and shielding that were employed. However, it was still possible to quantify 1.7 x 10^3 Bq/g of ^{241}Am in the presence of 7.8 x 10^5 Bq/g of ^{137}Cs. Overall, detection limits for ^{241}Am, expressed as a Currie MDA and calculated using the algorithm designed for this purpose within CANBERRA *Genie* software, were typically less than 370 Bq/g. Figure 4 shows ^{241}Am MDAs as a function of ^{137}Cs activity concentration for the Tank W-1A project samples, and compares them to samples from two other projects. The comparison projects comprise:

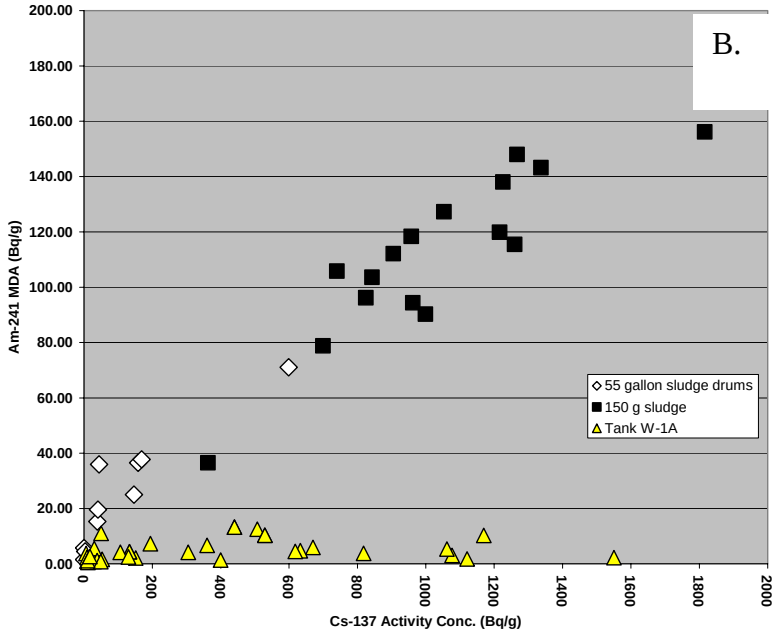
- 55 gallon drums containing sludge and characterized using 40% coaxial HPGe detectors
- 150 g sludge samples contained in 250 ml glass jars and characterized using a 50% broad energy germanium (BEGe) detector.

The Tank W-1A project utilized 50% BEGe detectors. A direct comparison with the 55 gallon drum and 150 g sludge projects is obviously not applicable due to the differences in detector type and sample matrix, and the complex differences in the details of the respective counting geometries. However, it is instructive to note that the apparatus used in the Tank W-1A project consistently provided ^{241}Am quantitative data that were useful to a TRU/LLW determination at ^{137}Cs levels where routine, less customized counting configurations might have been expected to fail.

CONCLUSIONS

On-site high-resolution gamma spectroscopy of soil cores was an effective tool for quantifying the three dimensional spatial extent of ^{241}Am, ^{137}Cs, and other gamma-emitting contaminants in the vicinity of Corehole 8 Tank W-1A. Soil cores were measured, analyzed, and reported with same-day turnaround times to support the dynamic Sampling and Analysis Plan. Special shielding geometries were developed and deployed to facilitate the detection and quantification of low and moderate concentrations of ^{241}Am in the presence of very high concentrations of ^{137}Cs. Two-dimensional spatial maps of the extent of the ^{137}Cs and ^{241}Am contamination were generated based on the NDA results. A conservative scaling factor of 5.0 was applied to the ^{241}Am results to generate a two-dimensional map of the estimated extent of the total TRU contamination. The estimated extent of the total TRU contamination was approximately 44m^2.


A.
Am-241 MDA (Bq/g)
Cs-137 Activity Conc. (Bq/g)
55 gallon sludge drums
Tank W-1A


B.
Am-241 MDA (Bq/g)
Cs-137 Activity Conc. (Bq/g)
55 gallon sludge drums
150 g sludge
Tank W-1A

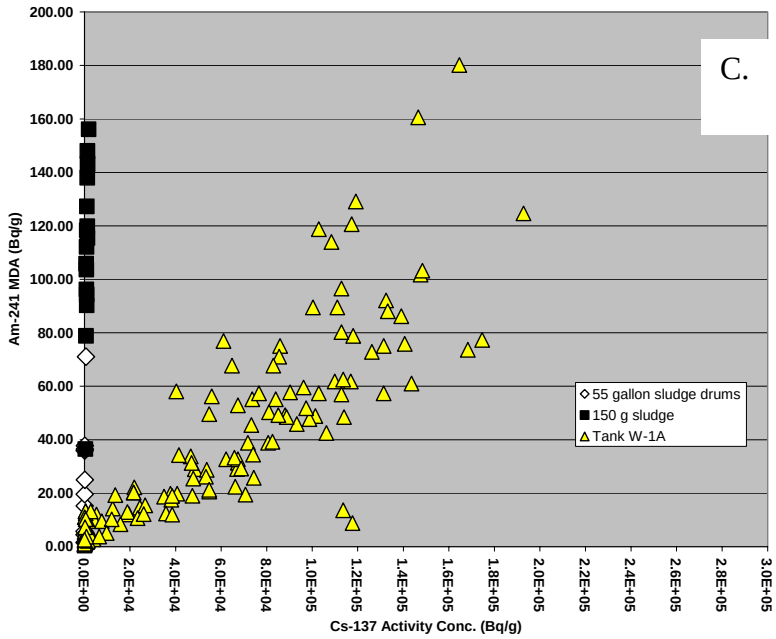


Figure 4. Am-241 minimum detectable activities (MDAs) as a function of Cs-137 levels in three projects. A, B and C focus upon different ranges of Cs-137 activity concentration.

REFERENCES

1 *Removal Action Report for the Core Hole 8 Plume Source (Tank W-1A) at the Oak Ridge National Laboratory*, Oak Ridge, Tennessee, DOE/OR/01-1969&D1, Bechtel Jacobs Company LLC, 2001.

2 *Sampling and Technology Selection Plan for the Core Hole 8 (Tank W-1A) TRU Soil Engineering Study for Completion of Removal Activities*, Oak Ridge National Laboratory, Oak Ridge, Tennessee, BJC/OR-2214, Oak Ridge, TN.

3 *Sampling and Analysis Plan for the Core Hole 87 (Tank W-1A) TRU Soil Engineering Study for Completion of Removal Activities*, Oak Ridge National Laboratory, Oak Ridge, Tennessee, BJC/OR-2215, Oak Ridge, TN.